MAY 2019

Argonne NATIONAL LABORATORY

2019 APS/CNM USERS MEETING

Program and Abstracts

2019 APS/CNM USERS MEETING

PROGRAM AND ABSTRACTS

User Facilities at Argonne National Laboratory

User Contacts

Advanced Photon Source
http://www.aps.anl.gov
630-252-9090
apsuser@aps.anl.gov

Argonne Leadership Computing Facility
http://www.alcf.anl.gov
630-252-0929

Argonne Tandem Linac Accelerator System
http://www.phy.anl.gov/atlas
630-252-4044

Center for Nanoscale Materials
http://nano.anl.gov
630-252-6952
cnm_useroffice@anl.gov

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The 2019 APS/CNM Users Meeting theme recognizes 2019 as the International Year of the Periodic Table of Chemical Elements.

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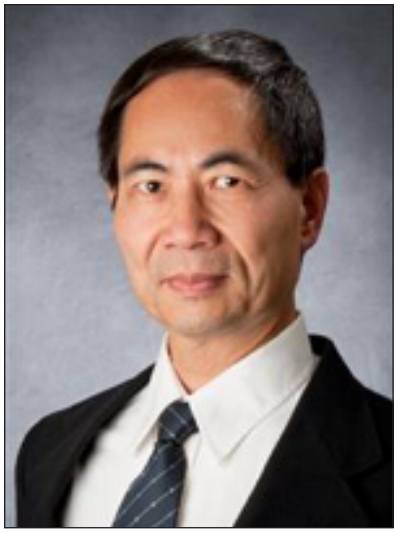
About Argonne National Laboratory

Argonne is a U.S. Department of Energy laboratory managed by the UChicago Argonne, LLC under Contract No. DE-AC02-06CH11357. The Laboratory is located southwest of Chicago at 9700 South Cass Avenue, Lemont, Illinois 60439. For information about Argonne, see www.anl.gov.



2019 APSUO Compton Award
Prof. Tai-Chang Chiang
(University of Illinois at Urbana-Champaign)

Professor Tai-Chang Chiang, Emeritus and Research Professor of Physics at the University of Illinois at Urbana-Champaign (UIUC), has been chosen to receive the 2019 Arthur H. Compton Award, presented by the Advanced Photon Source Users Organization (APSUO), "for his ingenuity and insight in developing x-ray thermal diffuse scattering into an efficient quantitative method for phonon band structure studies."

Beginning in the 1980s, Prof. Chiang was a pioneer in the use of synchrotron radiation in establishing the angle-resolved photoemission technique for electron band structure determination of solids, surfaces, and films.

His more recent scientific accomplishment is in developing the thermal diffuse scattering (TDS) technique into an efficient, quantitative method by which materials scientists at synchrotron x-ray facilities such as the Advanced Photon Source (APS) can determine phonon band structure. The impact of this methodology on materials research, especially in cases involving small samples, microstructures, and phase transitions is likely to be far reaching.

The TDS scattering patterns, recorded with a two-dimensional detector, show intricate details involving phonons from a wide range in k space over many zones. For instance, a least-squares analysis of the data acquired in just 20 seconds at APS Sector 33 yielded phonon dispersion curves of silicon over the entire Brillouin zone, which are in excellent agreement with neutron scattering results. This work was cited by Prof. Millie Dresselhaus in a plenary talk at the 2000 Fall MRS Meeting as one of three major achievements from all U.S. Department of Energy (DOE) facilities. The results were adopted by Jens Als-Nielsen and Des McMorrow in their textbook, *Elements of Modern X-Ray Physics* (Wiley, 2001), pg. 141.

Chiang's group has since extended the work to a variety of systems and phenomena. Particularly notable research includes phonon softening in the charge-density-wave compound TiSe2; phonon anomalies in niobium and plutonium; renormalization and fluctuation effects associated with the antiferrodistortive transition in $SrTiO_3$; and a new method of direct data inversion based on momentum-resolved x-ray calorimetry.
Ian K. Robinson
Professor of Physics, University College London

Over the course of his career, Prof. Chiang has made lasting contributions to condensed matter physics, surface science, and synchrotron radiation research, including several truly groundbreaking findings. He has authored some 300 journal articles, and his work has been cited more than 8500 times.

Chiang's work on precision electron spectroscopy and structural determination has led to important advances in the physics of electrons, lattice structures, and phonons and their mutual interactions in solids, at surfaces, in films, and at the nanoscale.

In 2016, he was elected by the Academia Sinica to its 2016 class of Academicians. He was among 22 scholars across all academic disciplines to receive that high honor that year. Academia Sinica is the national academy of Taiwan.

Chiang is the recipient of numerous other recognitions, including the 2015 Davisson-Germer Prize in Atomic or Surface Physics, sponsored by the American Physical Society. He received the Xerox Award for Faculty Research (1985), the National Science Foundation Presidential Young Investigator Award (1984-89), and the IBM Faculty Development Award (1984-5). He is a Fellow of the American Physical Society.

He served as head of the UIUC Solid State Sciences and Materials Chemistry Program from 1991 to 2006. He was associate director of the Frederick Seitz Materials Research Laboratory at UIUC from 1999 to 2006. From 2003 to 2008, he chaired the Board of Governors for the University-National Laboratory-Industry Collaborative Access Team at the Advanced Photon Source. From 2010 to 2014, he served as the scientific director of the University of Wisconsin-Madison Synchrotron Radiation Center. He was appointed Chair Professor at the National Chiao-Tung University at Taiwan (2013-16); Honorary Chair at National Tsing Hua University at Taiwan (2008-11); and Distinguished Chair at National Taiwan University (2007-10 and 2015-present). He is currently a visiting professor at Tokyo University.

Prof. Chiang received a bachelor's degree in physics from the National Taiwan University in 1971 and a doctoral degree in physics from the University of California, Berkeley, in 1978. He held a postdoctoral appointment at the IBM T.J. Watson Research Center in Yorktown Heights from 1978 to 1980, before he joined the faculty in the Department of Physics at the University of Illinois Urbana-Champaign in 1980.

About the Award

The Arthur H. Compton award was established in 1995 by the APS Users Organization to recognize an important scientific or technical accomplishment at the Advanced Photon Source. The award consists of a plaque and $2500.

The bi-annual awards are presented at the APS/CNM User Meetings, which are held every spring. A call for nominations is sent out before the meeting, and the winner(s) is invited to give an award lecture at the meeting. Awards are not necessarily made each year.

Compton was an American physicist who won the Nobel Prize for Physics in 1927 for discovering and explaining changes in x-ray wavelengths resulting from x-ray collisions with electrons, the so-called Compton effect. This important discovery in 1922 confirmed the dual nature (wave and particle) of electromagnetic radiation. A Ph.D. from Princeton University, Compton held many prominent positions, including professor of physics at The University of Chicago and chairman of the committee of the National Academy of Sciences that studied the military potential of atomic energy. His position on that committee made Compton instrumental in initiating the Manhattan Project, which created the first atomic bomb.

The Advanced Photon Source is a U.S. DOE Office of Science User Facility operated for the DOE Office of Science by Argonne National Laboratory under Contract No. DE-AC02-06CH11357.

Argonne National Laboratory seeks solutions to pressing national problems in science and technology. The nation's first national laboratory, Argonne conducts leading-edge basic and applied scientific research in virtually every scientific discipline. Argonne researchers work closely with researchers from hundreds of companies, universities, and federal, state and municipal agencies to help them solve their specific problems, advance America's scientific leadership and prepare the nation for a better future. With employees from more than 60 nations, Argonne is managed by UChicago Argonne, LLC, for the U.S. DOE Office of Science.

The U.S. Department of Energy's Office of Science is the single largest supporter of basic research in the physical sciences in the United States and is working to address some of the most pressing challenges of our time. For more information, visit the **Office of Science website**.

Previous award recipients

Nikolai Vinokurov and Klaus Halbach (1995)

Philip M. Platzman and Peter Eisenberger (1997)

Donald H. Bilderback, Andreas K. Freund, Gordon S. Knapp, and Dennis M. Mills (1998)

Sunil K. Sinha (2000)

Wayne A. Hendrickson (2001)

Martin Blume, L. Doon Gibbs, Denis McWhan, and Kazumichi Namikawa (2003)

Günter Schmahl and Janos Kirz (2005)

Andrzej Joachimiak and Gerold Rosenbaum (2007)

Gerhard Grübel, Simon Mochrie, and Mark Sutton (2009)

Edward Stern, Farrel Lytle, Dale Sayers (posthumously), and John Rehr (2011)

David E. Moncton, John N. Galayda, Michael Borland, and Louis Emery (2013)

Gene E. Ice, Bennett C. Larson, and Cullie J. Sparks (2015)

COMPREHENSIVE PROGRAM

Monday, May 6

8:00 – 5:00 Exhibits
Building 402, Gallery (lower level), outside E1100/1200 and Building 402, Atrium

7:30 – 5:00 Registration
Building 402, Atrium

12:00 – 1:30 Lunch (in the Tent)

Opening Session—Morning
Building 402, Lecture Hall

Session Chairs **R. Joseph Kline, R. Joseph Kline, Leader of Dimensional Metrology for Nanofabrication Project, National Institute of Standards and Technology and Olga Makarova, Creatv MicroTech, Inc.**

8:30 – 8:35 R. Joseph Kline, Chair, APS Users Organization
and Olga Makarova, CNM Users Executive Committee
Welcome and Launch of the 2019 Meeting

8:35 – 8:45 Paul K. Kearns, Argonne Laboratory Director
Welcome from the Laboratory

8:45 – 9:10 Bruce Garrett, Director of the Chemical Sciences, Geosciences, and Biosciences Division, Basic Energy Sciences, DOE
The DOE Perspective

9:10 – 9:25 Dennis Mills, Member of the Board of Directors for the Society for Science at User Research Facilities (SSURF)
Update on the Society for Science at User Research Facilities

9:25 – 9:30 Stephen Streiffer, APS Director
Introduction of Keynote Speaker

9:30 – 10:10 Keynote: Dan Nocera (Harvard University)
APS Insights to Metal-Oxygen and Metal-Halide Bond Activation for Energy Storage

10:10 – 10:35 Break

10:35 – 10:50 Stephen Streiffer, APS Director
APS Update

10:50 – 11:05 Supratik Guha, CNM Director
CNM Update

11:05 – 11:20 Robert O. Hettel, APS Upgrade Director
APS Upgrade Update

11:20 – 11:25 R. Joseph Kline, Chair, APS Users Organization
and Olga Makarova, CNM Users Executive Committee
Introduction of the Speed Science Slam

11:25 – 11:45 S^3: Speed Science Slam
APS: Roman Ezhov (Purdue University)
New Pathways in Iron-based Water Oxidation Catalysis

CNM: Tejas Guruswamy (Argonne National Laboratory)
Hard X-ray Transition Edge Sensors at the Advanced Photon Source

APS: Prabhat KC (Argonne National Laboratory and The University of Chicago)
Convolutional Neural Network Based Super Resolution for X-ray Imaging

CNM: Dali Sun (North Carolina State University)
Spintronic Terahertz Emission by Ultrafast Spin-charge Current Conversion in Organic-inorganic Hybrid Perovskites/Ferromagnet Heterostructures

11:45 – 12:00 Visit with Exhibitors

12:00 – 1:30 Lunch (in the Tent)

Parallel Facility Plenary Sessions—Afternoon

APS Session
Building 402, Lecture Hall

Session Chair: Carlo Segre, Illinois Institute of Technology

1:30 – 1:50 Oliver Schmidt (Argonne National Laboratory)
The APS Upgrade Beamlines Overview

1:50 – 2:30 2019 Arthur H. Compton Award Presentation and Talk:
Tai-Chang Chiang (University of Illinois, Urbana-Champaign)
X-ray Thermal Diffuse Scattering: History, Advances, and Opportunities

2:30 – 3:10 Yulia Pushkar (Purdue University)
X-ray Imaging and Spectroscopy of the Brain

3:10 – 3:25 Break

3:25 – 3:40 Student Invited Talk: Saugat Kandel (Northwestern University)
On the Use of Automatic Differentiation for Phase Retrieval

3:40 – 4:20 Melissa Sims (Johns Hopkins University)
Simulating Meteor Impacts in the Diamond Anvil Cell

4:20 – 5:00 Lin Chen (Argonne National Laboratory and Northwestern University)
Peeking into Excited State Trajectories with Pulsed X-rays: A Journey of Two Decades

5:00 Adjourn

6:00 Banquet
“Synchro De Mayo”
Argonne Guest House and Patio

Parallel Facility Plenary Sessions—Afternoon

CNM Session
Building 402, Room A1100

Session Chair: James Rondinelli, Northwestern University

1:30 – 2:15 Keynote Speaker: Ali Yazdani (Princeton University)
Visualizing Novel Quantum States of Matter

2:15 – 3:00 Paul Evans (University of Wisconsin at Madison)
Expanding the Scope of Nanobeam Diffraction: Dynamical Diffraction in Epitaxial Electronic Materials

3:00 – 3:20 Break

3:20 – 3:30 James Rondinelli, Chair, CNM Users Executive Committee (Northwestern University)
Update from the CNM Users Executive Committee

3:30 – 4:00 Hui Claire Xiong (Boise State University)
Development and Understanding of the Origin of Irreversibility in Layered Transition-metal Oxide Cathode Material for Sodium Ion Batteries

4:00 – 4:30 Peijun Guo (Argonne National Laboratory)
Hybrid Organic-inorganic Perovskites: From Anisotropic Excitons to Soft Crystal Lattice

4:30 – 4:45 Student Invited Talk: Frank Barrows (Northwestern University)
Fabrication, In Situ *Biasing, Electron Holography, and Elemental Analysis of Patterned and Unpatterned TiO_2 Thin Films*

4:45 Adjourn

6:00 Banquet
"Synchro De Mayo"
Argonne Guest House and Patio

Tuesday, May 7

8:00 – 5:00 Exhibits
Building 402, Gallery (lower level), outside E1100/1200 and Building 402, Atrium

8:00 – 5:00 Registration
Building 402, Atrium

8:30 – 12:00 Facility Specific Workshops

12:00 – 2:00 Poster Setup
(shuttle buses and vans provided throughout the lunch hour to provide transportation between APS, the Guest House, and TCS Building 240)

12:00 – 1:30 Lunch (in the Tent)

1:30 – 5:00 Facility Specific Workshops

5:30 – 8:00 Poster Session
TCS Building 240, Shuttle service available

Parallel Facility-specific Workshops

APS/CNM Workshop 1 – Building 402, Room E1100/1200
Driving Scientific Discovery with Artificial Intelligence, Advanced Data Analysis (see page 25)

APS Workshop 3 – Building 401, A1100
Workshop on Chemical Separations (see page 35)

CNM Workshop 4 – Building 401, Room A5000
Photon Qubit Entanglement and Transduction (see page 42)

APS Workshop 7 – Building 446, Conference Room
In Situ *and Multimodal Microscopy for APS-U (see page 57)*

Wednesday, May 8

8:00 – 1:30 Exhibits
Building 402 Gallery, outside E1100/1200 and Building 402 Atrium

8:00 – 1:30 Registration
Building 402, Atrium

8:30 – 12:00 Facility Specific Workshops

12:00 – 1:30 Lunch (in the Tent)

1:30 – 5:00 Facility Specific Workshops

Parallel Facility-specific Workshops

APS/CNM Workshop 2 – Building 401, Room A1100
Topological Quantum Information Science (see page 30)

APS Workshop 5 – Building 402, Room E1100/1200
RIXS after the APS Upgrade: Science Opportunities (see page 47)
Sponsored by Kohzu

CNM Workshop 6 – Building 446, Conference Room
Optoelectronic Devices and Mechanical Systems Based on Ultra-thin 2D Materials (see page 52)

Thursday, May 9

CNM Short Courses

8:30 – 12:00 CNM Short Course A: Building 440, Room B108 (attendees should meet in the CNM Lobby)
Introduction to Confocal Raman Spectroscopy
Instructor: Dave Gosztola (CNM)

A hands-on demonstration of the capabilities of the CNM's confocal Raman microscopes will be presented. Subjects to be discussed and demonstrated include the following:

- Basic Raman spectroscopy concepts
- Anatomy of a confocal Raman microscope
- Sample preparation
- Simple spectra collection
- Effects of excitation wavelength
- Point mapping
- Line/area mapping
- Heating and cooling samples

1:00 – 4:30 CNM Short Course B: Building 440, Room A105/106
Use of Machine Learning in Nanoscale Materials Modeling
Special Requirement: Bring your own laptop
Instructors: Maria Chan and Subramanian Sankaranarayanan (CNM)

This short course will include instructions on the use of machine learning for (1) force field parameterization from first principles data, and (2) nanoscale structure determination from a combination of modeling and experimental data. Experimental and computational users are encouraged to attend. For hands-on work, participants should be comfortable using a Linux command line environment and should bring their own laptop. Participants are encouraged to bring their research problems for diagnostics.

9:00 – 3:00 CNM Short Course C: Building 212, Room A157 and Building 212, D-Wing Microscopy Labs
Transmission Electron Microscopy (TEM) for Materials Science
Instructors: Yuzi Liu and Jianguo "JG" Wen (CNM)

This course targets the researcher with an entry to medium level knowledge of transmission electron microscopy (TEM). (S)TEM characterization and Aberration-corrected S/TEM for advanced energy material techniques will be introduced along with real examples. A demo of these techniques will follow the presentation.

APS Satellite Workshops

8:00 – 4:30 APS Satellite Training Course: Building 401, Room A5000

SAXS Software Software Packages Irena and Nika (Day 1)

Instructor: Jan Ilavsky (Argonne National Laboratory)

Successful small-angle x-ray scattering (SAXS, SANS) experiment requires appropriate data reduction and analysis tools. Igor Pro based packages Nika (for SAXS data reduction) and Irena (for SAS = SAXS/SANS/USAXS/USANS data analysis) were developed during the last 15 years at the APS. They are already being used widely for material science SAS at the APS and at other facilities worldwide. These tools are commonly included in syllabus of "Beyond Rg Materials" - SAXS short course organized semi-annually by APS SAXS SIG. However, the main audience of this SAXS short course is new SAXS users, starting with their own SAXS program and therefore it focuses more heavily on experiments, theory etc. The time devoted in this course for software is insufficient for experienced experimenters interested in complex software applications.

Therefore, the APS SAXS Special Interest Group (SIG) is organizing this specialized, hands-on, course specifically on the SAS software Nika and Irena. The course will be taught by the software author, Jan Ilavsky, APS staff member. In order to provide high educational value, the number of participants will be limited to 20. Participants are expected to have a high level of SAS experience and bring their own computers (Windows or OSX). In addition, they are encouraged to bring their own SAS experimental results.

Daily Schedule

8:00 Welcome coffee and continental breakfast

8:30 Course begins

~10:30 Break

12:00 Lunch (1 hour on own)

~2:30 Break

4:30 Adjourn

Working Agenda: Thursday, May 9

1. Installation of both packages/verification of functionality
2. Nika data reduction:
    a. Orientation in the program
    b. Calibration/geometry
    c. Mask design
    d. Data processing, sector and circular data reduction
    e. GI geometry, line profile, profile around ellipse…
    f. Programing for Nika (look up functions)
3. Irena data analysis:
    a. Orientation in the program

8:15 – 4:00 APS Satellite Training Course: Building 401, Room A1100

XANES and High-energy Resolution Fluorescence Detection (HERFD) XANES

Instructor: Matt Newville (GSECARS) and Josh Kas (University of Washington)

This training course will provide a description of measurement methods for high-resolution XANES and on data processing and analysis methods of XANES and resonant inelastic scattering. The course includes a mixture of practical training of analysis methods with talks on applications and advanced methods.

Daily Schedule

8:15 Welcome coffee and continental breakfast

9:00 Course begins

~10:30 Break

12:00 Lunch (1 hour on own)

~2:30 Break

4:00 Adjourn

Morning Session: Empirical Analysis for XANES + Methods
(Matthew Newville, University of Chicago)

1. Introduction, Data Processing, Over-absorption corrections.
2. Principal Component Analysis
3. Linear combination fitting
4. Multivariate / dimensionality reduction with LASSO-based supervised learning.
5. Fitting Pre-edge peaks

Afternoon Session: Theoretical Simulations of XANES + Methods
(Josh Kas, University of Washington)

Friday, May 10

APS Satellite Workshops

8:00 – 4:30 APS Satellite Training Course: Building 401, Room A5000
SAXS Software Software Packages Irena and Nika (Day 2)
Instructor: Jan Ilavsky (Argonne National Laboratory)

Daily Schedule

8:00 Welcome coffee and continental breakfast

8:30 Course begins

~10:30 Break

12:00 Lunch (1 hour on own)

~2:30 Break

4:30 Adjourn

Working Agenda: Friday, May 10

3. Irena data analysis, continued:

    b. Import of data

    c. Data plotting

    d. Data manipulation

    e. Unified fit, advanced analysis

    f. Size distribution

    g. Modeling tools

    h. Scattering contrast calculator

    i. Analytical tools

    k. SA Diffraction tool

    l. Reflectivity tool

    m. other tools…until end of the day.

8:15 – 5:30 APS Satellite Training Course: Building 401, Room A1100

X-ray Absorption Spectroscopy Simulations using FDMNES

Organizers: Yves Joly (Institut Néel, CNRS, Grenoble, France) and Chengjun Sun (ANL)

8:15 Welcome coffee and continental breakfast

9:00 Lecture on X-ray Matter Interaction

9:45 Break

10:00 Application for XANES, X-ray Raman and Resonant Diffraction

11:00 Break

11:15 Presentation of the FDMNES Software

11:30 Practical – XANES

12:30 Lunch (on own)

2:00 Practical – Dichroism, Resonant Diffraction, Surface Resonant Diffraction, X-ray Raman

3:30 Break

3:45 Practical on Examples Proposed by the Participants

5:30 Adjourn

Participants must bring a laptop. A package including the software (Windows, Mac or Linux), documentation and set of examples will be provided. Software to plot spectra (Origin, Kaleidagraph, etc.) is required.

GENERAL SESSION ABSTRACTS

APS PLENARY

X-ray Thermal Diffuse Scattering: History, Advances, and Opportunities

Tai C. Chiang
Department of Physics, University of Illinois at Urbana-Champaign, Urbana, IL 61801

I will tell a story about my group's research program on utilizing thermal diffuse scattering (TDS) to investigate phonon properties of crystals. Right around the time the Advanced Photon Source opened its door to users, we were trying to perform surface diffraction with a transmission geometry, hoping to develop an efficient method for determining the atomic structure of reconstructed surfaces. Instead, we found beautiful TDS patterns from the bulk, which overwhelmed the surface signals. TDS arises from scattering by lattice vibrations (phonons). Debye realized in 1913 that lattice vibrations reduce the diffraction peak intensities (by the Debye Waller factor) and the lost intensities are dispersed into a diffuse but feature-rich TDS background. A number of pioneers in x-ray diffraction recognized that TDS could be useful for phonon studies, but early experiments based on x-ray tubes were inefficient and impractical. With the advent of neutron scatting in the 1950s, the x-ray TDS method was largely abandoned and forgotten. At APS, high-quality 2D TDS patterns over a wide range of momentum transfer could be acquired in seconds, but it took us quite a while to develop the mathematical tools needed to extract useful information from the data. I will show some examples to illustrate the technical progress and the utility of TDS for studies of phonons, soft modes, and phase transitions in various materials including tiny samples, which are difficult for neutron scattering. I will comment on what we are working on now, including time-resolved studies following pulsed excitations.

I wish to thank the people who made APS a reality and my group members, colleagues, the UNICAT team (with members from the University of Illinois, Oak Ridge, NIST, Allied Signal, and UOP), and APS staff who participated in and/or helped with this research.

APS PLENARY

X-ray Imaging and Spectroscopy of the Brain

Yulia Pushkar
Department of Physics, Purdue University, West Lafayette, IN 47906

The increasing prevalence of neurodegenerative diseases such as Alzheimer's (AD) and Parkinson's (PD) diseases is a major public health as well as economic concern in the USA and other developed countries. Currently, no treatment is available to slow the progression of these diseases and the aging population, which is disproportionately affected by these diseases, place an increasing strain on the economy. Recent research implicates trace elements (such as Mn, Fe, Cu and Zn) neurotoxicity in the etiologies of Alzheimer's (AD) and Parkinson's (PD) diseases but the molecular mechanisms are mostly unknown. The number of experimental techniques that can measure metal distribution and speciation in the brain is limited.

I will present x-ray fluorescence imaging, a method for quantitative analysis of elemental distribution in the brain; and x-ray diffraction imaging, which is sensitive to structural features in key elements of a healthy brain, such as myelin and neurofilaments—as well as pathological aggregates—such as plaques, Lewy bodies, and neurofiblillary tangles. Using XRF we studied Mn distribution in rat model of occupational Mn exposure [1–4]. We found that globus pallidus and substantia nigra compacta are areas in the brain that accumulate most Mn. Imaging the Mn distribution in dopaminergic neurons of exposed rats we determined that intracellular Mn range between 40–200 micromolar; concentrations as low as 100 micromolar have been observed to cause cell death. This is a first potentially direct link between Mn exposure and Parkinson's disease.

Via XRF analysis of healthy rodent brains have discovered localized Cu-rich aggregates in astrocytes of the subventricular zone with Cu concentrations in the hundreds of millimolar [5–7]. Based on a [S]/[Cu] ratio and x-ray absorption spectroscopy, metallothionein was proposed as a binding protein. An analysis of metallothionein(1,2) knockout mice by XRF shown decrease in number of Cu containing aggregates but these were not eliminated indicating alternative pathways for aggregates formation.

[1] Sullivan, B.; Robison, G.; Kay, M.; and Pushkar, Y. (2016). "Mn exposure does not affect neurogenesis," **152**(2): 257–261.

[2] Robison, G.; Sullivan, B.; Cannon, J.R.; and Pushkar, Y. (2015). "Identification of dopaminergic neurons of the substantia nigra pars compacta as a target of manganese accumulation," *Metallomics* **7**(5): 748–755.

[3] Robison, G.; Zakharova, T.; Fu, S.; Jiang, W.D.; Fulper, R.; Barrea, R.; Zheng, W.; and Pushkar, Y. (2013). "X-ray fluorescence imaging of the hippocampal formation after manganese exposure," *Metallomics* **5**(11): 1554–1565.

[4] Robison, G.; Zakharova, T.; Fu, S.; Jiang, W.; Fulper, R.; Barrea, R.; Marcus, M.A.; Zheng, W.; and Pushkar, Y. (2012). "X-ray Fluorescence Imaging: A New Tool for Studying Manganese Neurotoxicity," *PLoS One* **7**(11).

[5] Sullivan, B.; Robison, G.; Pushkar, Y.; Young, J.K.; and Manaye, K.F. (2017)."Copper accumulation in rodent brain astrocytes: A species difference," **39**: 6–13.

[6] Sullivan, B.; Robison, G.; Osborn, J.; Kay, M.; Thompson, P.; Davis, K.; Zakharova, T.; Antipova, O.; and Pushkar, Y. (2017). "On the nature of the Cu-rich aggregates in brain astrocytes," **11**: 231–239.

[7] Pushkar, Y.; Robison, G.; Sullivan, B.; Fu, S.X.; Kohne, M.; Jiang, W.; Rohr, S.; Lai, B.; Marcus, M.A.; Zakharova, T.; and Zheng, W. (2013). "Aging results in copper accumulations in glial fibrillary acidic protein-positive cells in the subventricular zone," *Aging Cell* **12**(5): 823–32.

APS PLENARY

On the Use of Automatic Differentiation for Phase Retrieval

Saugat Kandel[1], S. Maddali[2], Ming Du[3], Marc Allain[4], Stephan O. Hruszkewycz[2], Chris Jacobsen[5,6,7], and Youssef Nashed[8]

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The recent rapid development in coherent diffraction imaging (CDI) methods has enabled nanometer-resolution imaging in a variety of experimental modalities. Image reconstruction with such CDI methods involves solving the phase retrieval problem, where we attempt to reconstruct an object from only the amplitude of its Fourier transform. This can be framed as a nonlinear optimization problem which we can solve using a gradient-based minimization method. Typically, such approaches use closed-form gradient expressions. For complex imaging schemes, deriving this gradient can be difficult and laborious. This restricts our ability to rapidly prototype experimental and algorithmic frameworks.

In this work, we use the *reverse-mode automatic differentiation* method to implement a generic gradient-based phase retrieval framework. With this approach, we only need to specify the physics-based forward propagation model for a specific CDI experiment; the gradients are exactly calculated automatically through a sequential application of the chain rule in a *reverse pass* through the forward model. Our gradient calculation approach is versatile and can be straightforwardly implemented through various deep learning software libraries (TensorFlow, Pytorch, Autograd, etc.), allowing for its use within state-of-the-art accelerated gradient descent algorithms. We demonstrate the generic nature of this phase retrieval method through numerical experiments in the transmission (far-field and near-field), Bragg, and tomographic CDI geometries.

APS PLENARY

Simulating Meteor Impacts in the Diamond Anvil Cell

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Impact cratering is important in planetary body formation and evolution [1,3]. The pressure and temperature conditions reached during impacts are classified using systems [4–6] that stem from 1) petrographic deformation features observed in the impactite and 2) the presence of high pressure mineral phases. However, the effects of kinetics and strain-rate on deformation features and high-pressure phases are relatively unconstrained [7]. We completed room temperature, 300°C, and laser-heated rapid compression and decompression membrane and dynamic diamond anvil cell (mDAC and dDAC) experiments with *in situ* x-ray diffraction [2]. We studied olivine ($(Mg, Fe)_2 SiO_4$) and plagioclase ($NaAlSi_3O_8$-$CaAl_2Si_2O_8$). The compression and decompression rates were between 0.04–81.0 GPa/s up to pressures around 50 GPa. The compression rates allow us to observe deformation and transformation mechanisms as well as thermodynamic effects in detail. In plagioclase, we identify a specific mechanism for amorphization and define decompression effects on nucleation. In olivine, we suggest what mechanisms lead to the deformation features observed in shocked samples. These experiments have allowed us to identify some of the complex interactions that likely play a role in natural events and are previously unexplored.

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APS PLENARY

Peeking into Excited State Trajectories with Pulsed X-rays: A Journey of Two Decades

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I will give a very brief reflection of 20 years in taking snapshots of photoexcited molecular species in solar energy conversion processes at the APS and beyond. Then I will use two recent examples to demonstrate how x-ray spectroscopies from the APS can decipher both electronic and nuclear geometry in the ground and excited state molecular species.

Electronic Structures of Metal Centers in an OER Catalyst Model. One of the challenges in solar fuels generation is generating multiple redox equivalents for the oxygen evolution reaction (OER) (i.e., $2H_2O \rightarrow O_2+4H^++4e^-$) that is a four-electron, four-proton coupled process. In collaboration with Nocera Group, we used a combination of *in situ* and *ex situ* x-ray absorption and emission spectroscopies as well as quantum mechanical calculations on OER thin films and their molecular and heterogeneous inorganic analogs. These studies provided insights into the electronic structures of the high-valent states involved in the mechanisms of O-O bond formation and for the high-valent states to drive the bond-forming and bond-breaking steps of OER reactions. We employed complementary x-ray spectroscopies, x-ray absorption (XAS) and 1s3p resonant inelastic x-ray scattering (K_β RIXS), to effectively extract Co(IV) contributions within a spectroscopically active background. Co K- and L-edge x-ray absorption directly probe the 3d-manifold of effectively localized Co(IV), providing a handle on the covalency of the dπ-based redox active molecular orbital (MO) of Co_4O_4. We also probed the doubly oxidized $Co(III)_2(IV)_2$ state of the Co_4O_4 cluster, which features a cofacial $Co(IV)_2$ site and is thus a molecular model of the active site in Co-OEC.

Electron and Energy Relays in the Excited State Supramolecular Dinuclear Transition Metal Complexes. The rational design of multinuclear transition metal complexes (TMCs) for photochemical catalysis of homogeneous and/or heterogeneous multi-electron reactions (e.g., for producing solar fuels) requires a detailed understanding of the often unique and convoluted excited state charge and energy transfer dynamics in this class of compounds. Using combined optical and x-ray transient spectroscopic measurements, excited state electron electron relays in tetrapyridophenazine-bridged heteroleptic dinuclear Cu(I) bis(phenanthroline) complexes have been investigated as an exemplary system.

CNM PLENARY

Visualizing Novel Quantum States of Matter

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Spectroscopic mapping with the scanning tunneling microscope (STM) at low temperatures allows us to explore quantum materials in an unprecedented fashion. In this talk, I will describe a few studies at the forefront of research in quantum condensed matter physics. One area of research is related to topological quantum computing in which we have used the STM to visualize an exotic quasi-particle called a Majorana zero mode. I will also describe STM studies in very high magnetic field in which we provide direct visualization of quantized electrons' Landau orbits, which that underlie the formation topological quantum Hall state. I will close with an outlook of some of the exciting problems in the condensed matter physics, which can be addressed using STM spectroscopic mapping techniques.

CNM PLENARY

Expanding the Scope of Nanobeam Diffraction: Dynamical Diffraction in Epitaxial Electronic Materials

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Epitaxial electronic materials include a thin film or nanostructure that shares key aspects of its crystallographic structure with a thick underlying substrate. In key applications of these epitaxial materials in the development of semiconductor quantum computing, the control of this structure at the scale of tens to hundreds of nanometers has been an important challenge. Semiconductor quantum computing devices based on heterostructures formed in the AlGaAs/GaAs system involve thin epitaxial layers with tiny lattice mismatches

on the order of 10^{-4} or smaller and rely on the formation of qubits defined at the lateral scale of tens of nanometers. The further mechanical distortion of these layers during the formation of devices causes significant electronic perturbations due to the piezoelectric and deformation potential effects. Ultimately, the mechanical distortion and the accompanying electronic perturbation affects the operation of devices. It has been challenging, however, to characterize the mechanical distortion of these epitaxial layers, in part because the close lattice match between the substrate and the thin film has posed a difficult problem for x-ray nanobeam diffraction techniques. We have found that this problem can be addressed through the development of new nanobeam diffraction methods in which dynamical diffraction simulations are employed to interpret the complex diffraction patterns arising from latticed-matched heterostructures [1].

Nanobeam diffraction studies of lattice-matched GaAs/AlGaAs quantum computing devices reveal mechanical distortions due to device fabrication, including from the residual stress and thermal expansion coefficient mismatch associated with metallic electrodes [2,3]. This new understanding the magnitude and role of these effects has the potential to allow the design of improved device geometries and potentially to lead to devices in which strain, in addition to electrostatic effects, is used to define quantum devices.

Beyond quantum computing structures, similar dynamical diffraction effects appear in problems associated with ferroelectric domains and domain boundaries in epitaxial $BaTiO_3$ thin films and in strain sharing effects in nanoscale complex oxides [4].

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CNM PLENARY

Development and Understanding of the Origin of Irreversibility in Layered Transition-metal Oxide Cathode Material for Sodium Ion Batteries

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Sodium ion batteries are attractive alternative energy storage technology to lithium-ion batteries due to its low-cost. There has been growing attention in developing new cathode materials for sodium ion batteries. The Iron-based layered oxide cathode is of significant interest due to the low-cost, abundant, environmentally-friendly material selection. The O3-type layered $Na(Ni_xFe_yMn_z)O_2$ ($0 < x, y, z < 1$) (NFM) cathode materials have attracted great interest in sodium-ion batteries due to the abundance and cost of raw materials and their high specific capacities. However, the cycling stability and rate capability at high voltages (> 4.0V) of these materials remains an issue. In this work, we investigated the effect of Li substitution in NFM cathode and successfully synthesized a Li-substituted layered-tunneled (O3-spinel) intergrowth cathode (LS-NFM) to address these issues.

The remarkable structural compatibility and connectivity of the two phases were confirmed by x-ray diffraction (XRD), selected area electron diffraction (SAED) and high resolution transmission electron microscopy (HRTEM). The LS-NFM cathode exhibits enhanced cycling stability as well as rate capability compared to the un-doped NFM cathode. The enhanced rate capability of LS-NFM can be explained by the significantly increased effective Na^+ diffusivity measured by galvanostatic intermittent titration technique (GITT) compared to the un-doped control NFM cathode, which can be ascribed to the improved charge transport kinetics through shortened diffusion path by direct connection between the 3D channels in the spinel phase and 2D channels in the layered phase. The results from *ex situ* hard/soft x-ray adsorption spectroscopy (XAS) suggest that the capacity of LS-NFM cathode is mainly associated with the Ni^{2+}/Ni^{4+} redox couple, and slightly from the Fe^{3+}/Fe^{4+} redox couple. We also investigated the

origin of irreversibility of un-doped NFM cathode materials. It was found that the irreversibility became pronounced and the electrochemical performance became limited with the increase of either Fe composition or the upper cutoff potential in these materials.

CNM PLENARY

Hybrid Organic-inorganic Perovskites: From Anisotropic Excitons to Soft Crystal Lattice

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Hybrid organic-inorganic perovskites are solution-processed, scalable materials exhibiting remarkable optoelectronic properties such as strong light absorption, defect tolerance, and long carrier lifetimes. I will describe how electronic excitations in these materials are coupled to and influenced by the vibrational degrees of freedom of the organic and inorganic sublattices, investigated using an array of optical spectroscopic techniques. The unique soft nature of the lead-halide octahedral framework gives rise to dynamic fluctuations in the electronic bandgap, which distinguishes hybrid perovskites from traditional inorganic semiconductors. Furthermore, strong quantum confinement can be easily imparted to hybrid perovskites with the use of organic spacer-molecules, leading to exotic dispersion relation and enhanced light-emitting properties. Our results demonstrate that hybrid materials consisting of distinct sub-lattices can allow for dramatically enhanced light absorption, emission and charge carrier collection at various time- and length-scales.

CNM PLENARY

Fabrication, *in situ* Biasing, Electron Holography and Elemental Analysis of Patterned and Unpatterned TiO_2 Thin Films

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TiO_2 is a metal oxide that can undergo resistive switching, a reversible change between high and low resistance states by application of a voltage bias. This behavior has promising applications in neuromorphic computing and nonvolatile memory. In order to gain a deeper understanding of the mechanism behind reversible switching and electric breakdown in TiO_2 we have fabricated samples for *in situ* biasing and Transmission Electron Microscopy, (TEM). Additionally, we have patterned thin films of TiO_2 on the nanoscale as a means to gain additional insights into the reversible breakdown through *in situ* biasing.

I will present details of the fabrication process we developed at the Center for Nanoscale Materials, (CNM). This includes both the process to pattern TiO_2 thin films and the preparation of thin films for *in situ* biasing in TEM. In order to pattern TiO_2 , we perform sequential infiltration synthesis of Al_2O_3 into block copolymers to make a patterned film of Al_2O_3 on top of thin films of TiO_2. Using reactive ion etching we transfer the Al_2O_3 pattern into the TiO_2 thin film. To prepare these patterned and unpatterned samples for *in situ* biasing we use electron beam lithography to write electrodes on top of the TiO_2 thin films. Finally, we use wet etching to back etch SiN windows into our substrate. Additionally, I will present results from *in situ* biasing experiments. Using electron holography and electron energy loss spectroscopy (EELS) in the CNM, we have observed irreversible changes in our thin films during our biasing experiments. I will compare these results in the patterned and unpatterned TiO_2 films.

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